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Pressure Dependence of T_c in $Tl_2Ba_2CaCu_2O_8$
at Hydrostatic Pressures to 6 GPa

by

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Abstract

We report the results of experiments carried out on single crystals of $Tl_2Ba_2CaCu_2O_8$ under nearly hydrostatic pressure conditions to 6 GPa. The critical temperature, T_c , is observed to possess a non-linear pressure dependence. While the increase of T_c with increasing oxygen concentration in $YBa_2Ca_3O_{7-\delta}$ is consistent with the observation of increasing T_c with increasing pressure, the $Tl_2Ba_2CaCu_2O_8$ system demonstrates an increasing T_c with decreasing oxygen content, but has a positive dT_c/dP at low pressures. This behavior is clearly inconsistent with the picture applied to the YBCO system. In all the present measurements the curvature is downward and in fact for one sample it actually decreases. A theoretical explanation based upon the strong-coupling Eliashberg form of the BCS theory is put forth to explain this effect.

Studies of the pressure dependencies of the critical temperatures of hole-doped high T_c superconductors in the past have demonstrated, with very few exceptions, monotonically increasing T_c 's with increasing pressure.^{1,2} This is thought to be due to the pressure-induced injection of holes onto the conduction planes resulting from a change in the apical Cu-O bond length,^{3,4,5} a view consistent with the drop of critical temperature with decreasing oxygen concentration and hence decreasing hole concentration.⁶ Recently work has been performed which indicates that for many of the thallium-based cuprates, increases in the oxygen content in as-made samples, actually decreases T_c ,⁷ while decreases in oxygen content produce sharp increases in T_c , followed by decreases with further oxygen reduction.⁸ However studies of the pressure dependence of T_c have indicated a positive slope at low pressures.^{9,10,11} This effect is contrary to that observed for the YBCO systems.

In this *Letter*, we report the dT_c/dP results of experiments performed on several single crystals of $Tl_2Ba_2CaCu_2O_8$ in a nearly hydrostatic environment to pressures of 6 GPa utilizing *in situ* pressure measurement techniques at low temperature. Two previous experiments with sintered samples have indicated that the pressure dependence of T_c in the Tl "2212" compound, $Tl_2Ba_2CaCu_2O_8$, may not be monotonic but rather increases with increasing pressure at lower pressures, reaches a maximum, and then decreases with further increasing pressure.^{12,13} We also provide a theoretical explanation for the occurrence of the observed effects.

Samples were obtained from two preparation processes. Those in group A (samples later identified as having $T_c(0)$ of 112.1, 107.2, and 106.7 K) were grown

in sealed gold capsules using the off-stoichiometric flux method. The oxides Tl_2O_3 , BaO_2 , CaO_2 , and CuO were ground together for 45 min in a 2:2:3:4 molar metal ratio, respectively using an agate mortar and pestle. Approximately 3 g of the charge mixture was sealed in a gold capsule which was then heated under trapped flowing oxygen at 1 atm as follows: from ambient to 920 C in 1.5 h, soak 3 h, cool at 24 C/h to 750 C, furnace turned off and sample removed at room temperature. From the large number of black platelets embedded in a black polycrystalline matrix, an x-ray powder diffraction analysis was performed to determine the presence of the (2212) and (2223) phases in the approximate ratio of 60:40. Residual $BaCuO_2$ and CuO were clearly present.

The preparation of the group B ($T_c(0)$ of 114.1 K) began with appropriate amounts of Tl_2O_3 , BaO , CaO and CuO (each 99.99% pure), these were ground to form a powder with a nominal composition of $Tl_2Ba_2CaCu_2O_x$. The mixture was heated in air for 24 h at 920 C with an intermediate grinding carried out to facilitate a complete reaction. This powder was then pressed into a pellet under a load of 460 kg/cm². Pellets of mass 25 g were placed in an Al_2O_3 crucible, which was loosely covered by an Al_2O_3 plate to protect the Tl_2O_3 from direct evaporation. The crucible was then put into a quartz tube located in a horizontal tube furnace with the following schedule: a 15 min heating to 940 C - 945 C to melt most of the mixture, cooling at a rate 0.2 C/min to 900 C where crystals formed, cooling at 1 C/min to 700 C, 2 C/min to 400 C, and five more hours of furnace cooling to room temperature. This procedure relieved some of the inner stress and optimized the oxygen distribution and crystal structure. The x-ray dot maps and XPS show the crystals were chemically uniform with stoichiometries of (2212) and (2223) phase crystals. X-ray data recorded on a four circle diffractometer verified the (2212) single crystal was of high quality and no minor misoriented phase could be found.

In the (2223) phase crystals grown there are sometimes two or three subgrains with a few degrees of misorientation.

Pressure was generated in a diamond anvil cell¹⁴ employing both hardened Be-Cu 25 alloy and Inconel gaskets. Samples were small, laminar single crystals whose sizes varied, but were less than 350 μm on edge. Two different pressure transmitting fluids were used: light silicone diffusion pump oil for one sample and 4:1 methanol/ethanol for all the others studied. Pressure was measured using the ruby fluorescence technique^{15,16,17} *in situ* at about 100 K and temperatures were maintained with a helium transfer refrigerator modified for optical measurements. At pressures above 0.1 GPa, there was a significant increase in sample pressure upon cooling, clearly demonstrating the importance of the *in situ* pressure measurement. Temperatures were measured with a calibrated silicon diode mounted on the cell close to the sample.

Superconducting transitions were detected by AC susceptibility techniques. A pair of coils, with 50 turns on the primary and 250 turns on the secondary and each wound to a shape conforming to that of the diamonds, was mounted around the diamond attached to the fixed end of the cell. These were connected to a Hartshorn AC susceptibility bridge.¹⁸ Sinusoidal fields of approximately 0.2 Gauss rms were generated by a signal generator at about 2 KHz. Temperature sweeps were generally performed by warming from low temperatures at a rate of 1.0 ± 0.2 K/min. Controlled cooling sweeps demonstrated no hysteresis within the temperature resolution of the experiment. Both real and imaginary components of the susceptibility were detected with a lock-in amplifier. Figure 1 shows several transitions observed in the real susceptibility component for sample 1 at various pressures. A transition width of approximately 2 K is found between the 90 and 10

percent temperatures. And there is essentially no change in this width to the highest pressure.

The results of experiments conducted on four samples are shown in Figure 2. Sample 1 was a single crystal in a BeCu gasket with silicone oil for a pressure transmitting medium and had an ambient pressure onset temperature of 112 K. A large initial positive slope of about 4 K/GPa is indicated. At higher pressures, the slope decreased and at the highest pressure dropped from 122.7 K at 3.7 GPa, to 121.3 at 4.8 GPa. It was thought that this might arise from some effect of the non-hydrostaticity of the fluid, but examination of the ruby fluorescence spectra showed no observable broadening within the precision of the spectrometer. Samples 2, 3 and 4 were studied using inconel gaskets and 4:1 methanol/ethanol fluid. They had ambient pressure onset temperatures of 106.7 ± 0.4 K (extrapolated from a quadratic fit to high P data), 114.0 ± 0.2 K, and 107.2 ± 0.2 K, respectively. Each of these possesses a positive initial dT_c/dP . Values of 1.6 K/GPa, 2.0 K/GPa and 2.4 K/GPa were recorded for samples 2, 3 and 4 respectively and are consistent with previously reported values.^{9,10,11} At higher pressures these samples clearly demonstrate a downward curvature over the pressure range attainable with this system, although the maximum pressure was not high enough to observe any decrease. A similar effect has been observed recently by Mori et al.¹² and Allgeier et al.¹³ each using polycrystalline samples. Mori, et al. generated pressures with a cubic anvil device and with room temperature measurements of pressure based on bismuth fixed points. Allgeier et al. pressed the polycrystalline sample into a metal gasketed anvil cell but details of their pressure measurement were not given.

This remarkable pressure dependence can be explained in direct analogy with the dependence of T_c on the carrier concentration that is characteristic of many of the cuprate superconductors¹⁹ and particularly in the work of Martin et al.⁸ on the series of Tl cuprate superconductors with one, two and three Cu-O layers.

Recently Kresin and Moravitz²⁰ have explained the carrier dependence of the transition temperature in the cuprates, especially in the LaSrCuO "214" compound, by utilizing both the strong coupling BCS theory and the highly anisotropic quasi-2D Fermiology that exists because of the layered structure of the Cu-O planes. Using the strong-coupling Eliashberg form of the BCS theory, they find that T_c is a non-monotonic and very sensitive function of the Fermi wavevector k_F . This is due to the electron-phonon coupling parameter λ being proportional to k_F^2 when k_F is smaller than a limiting value of the phonon momentum, q_c , for the most important phonon mode, and proportional to $(k_F)^{-1}$ when $k_F \gg q_c$. Thus, as k_F crosses through q_c , the transition temperature is maximum and decreases on either side of this optimum condition. In a quasi 2D layered superconductor it has been shown that

$$(k_F)^2 = [2\pi n c]$$

where n is the carrier concentration and c is the distance between conducting 2D subsystems.²¹ Thus, not only is the transition temperature strongly affected by the carrier concentration, n , it is clearly just as strong a function of the interlayer spacing c . Therefore there should be a direct analogy between the dependence of T_c on n and on c .

The dependence on carrier concentration for the Tl "2212" compound has been measured by Martin et al.⁸ and Shimakawa et al.⁷ They clearly show that,

as made, this compound is "overdoped" as far as T_c is concerned. By a low temperature reducing anneal that removes oxygen and hence decreases n , Martin et al. found a transition temperature that rises very rapidly from about 96 K to a maximum of about 118 K and then starts to fall more slowly as the carriers are further removed. Thus k_F is larger than q_c to begin with and is reduced so that it crosses through q_c at some concentration giving rise to the non-monotonic dependence of T_c with carrier concentration. Similarly an increase of pressure reduces c and therefore k_F . The dependence of T_c on pressure should therefore mimic the dependence of T_c on n . The data in Fig. 2 are strikingly similar to the data in Table I in the Martin work. Of course for a quantitative comparison the Martin data would have to be plotted versus carrier concentration rather than the reducing time and the data presented here would have to be plotted versus c not pressure.²² It is also possible that pressure may indirectly affect carrier concentration in addition to its very direct affect on c . Therefore it may be very difficult to get quantitative agreement with the theory unless the carrier concentration is directly measured as a function of pressure as well as oxygen reduction.

An interesting test of the theory would be to perform first an oxygen reduction and then a determination of the pressure dependence of T_c . If k_F is reduced to a value less than q_c by reduction, further decreases in k_F resulting from decreases in c should result in a monotonically decreasing critical temperature and a negative value of dT_c/dP .

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Figure Captions:

Fig. 1: Offset voltage vs. sample temperature for $Tl_2Ba_2CaCu_2O_x$ single crystal for 5 pressures to 48 kbar.

Fig. 2: Increase in T_c with pressure for 4 different single crystal samples of $Tl_2Ba_2CaCu_2O_x$

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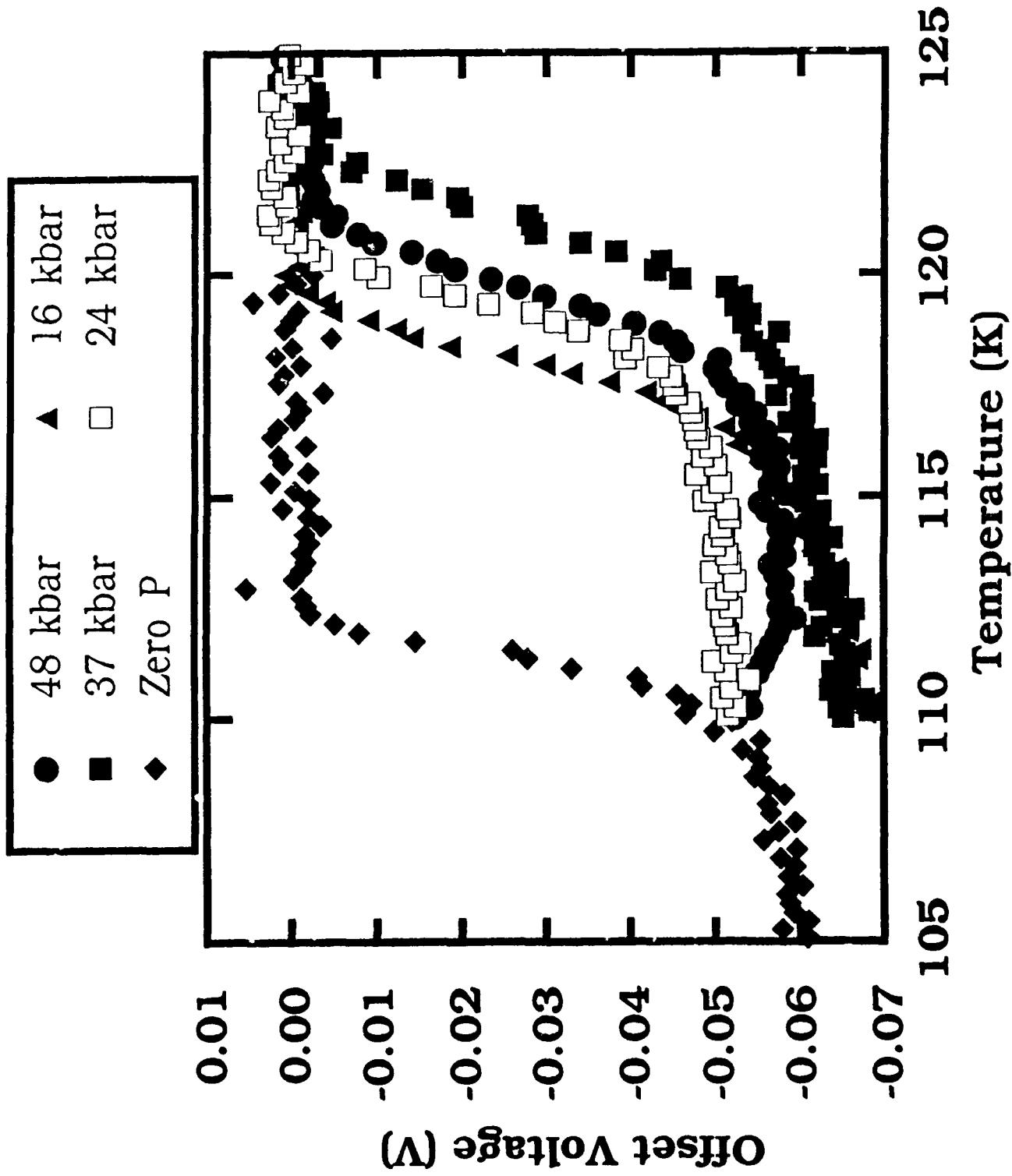
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- 22 Compressibility data have been collected recently from the same batch of samples used here at pressures to 50 GPa; these data are presently being analyzed in terms of the linear compressibilities and associated bulk moduli.



- ● - $T_c(0) = 112.1$ K
- ◆ - $T_c(0) = 106.7$ K
- ■ - $T_c(0) = 107.2$ K

